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Spin-glass transition in a model magnetic fluid: Electron spin resonance investigation of $Mn_{0.5}Zn_{0.5}Fe_2O_4$ nanoparticles dispersed in kerosene

R. V. Upadhyay, Kinnari Parekh, and R. V. Mehta
Laboratory of Ferrofluids, Department of Physics, Bhavnagar University, Bhavnagar-364 002, India

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A zero-field-cooled (ZFC) electron spin resonance (ESR) study of $Mn_{0.5}Zn_{0.5}Fe_2O_4$ nanoparticle-dispersed magnetic fluid exhibits an isotropic shift in resonance field below 50 K. Below this temperature the field-cooled ESR spectra (textured magnetic fluid) exhibits an angle-dependent hysteresis behavior in resonance field. The hysteresis effect diminishes above 60 K. In both the cases—i.e., ZFC and FC, the linewidth increases rapidly below 50 K. The results has been analyzed in light of random field exchange anisotropy model proposed by Martinez *et al.* Using this model the surface spin-glass layer thickness has been calculated. The value (3.8 Å) agrees well with that obtained from small-angle neutron scattering experiments (3 Å).

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I. INTRODUCTION

There is a great deal of evidence—like a peak in zero-field-cooled magnetization, high-field irreversibility in $M(H)$ curves, nonexponential relaxation behavior, etc., which indicate that a phase resembling a spin glass exists in a frozen nanomagnetic particle system.[1–4] Jonsson *et al.*[4] have shown that due to dipole-dipole interactions in a concentrated ferrofluid containing γ-Fe_2O_3 particles a ‘‘spin-glass-like’’ phase may occur at a low temperature. Martinez *et al.*[5] subsequently carried out magnetization measurement of γ-Fe_2O_3 nanoparticles in zero-field-cooled (ZFC) and field-cooled (FC) states. From these measurements it was concluded that there exists a surface spin-glass layer that freezes at $T_F \approx 42$ K. Using the random-field model of exchange anisotropy the thickness of this layer was calculated and was found to be 6 Å.[5] More recently, Mamiya *et al.*[6,7] have shown a critical slowing down of relaxation and a divergence of nonlinear susceptibility at a finite temperature in the frozen iron-nitride magnetic fluid. Effects of temperature changes on aging phenomena are interpreted using the droplet model. Kodama *et al.*[8] have also interpreted their observations of magnetization of $NiFe_2O_4$ nanoparticles at low temperatures in terms of a surface spin-glass layer. Thus it is now widely accepted that in a nano-magnetic-particle system there occurs a cooperative freezing of spins at a finite temperature T_g as in a normal spin glass. Though in several aspect this ‘‘spin-glass’’-like behavior is similar to that observed in conventional spin glass, but a fine-magnetic-particle system also behaves like a random anisotropy system unlike conventional spin glass.[3]

Magnetization and Mössbauer measurements are mostly employed in above studies. The electron spin resonance (ESR) technique has been used to study metallic spin glass, reentrant alloys, etc.[9–12] Its use in the study of magnetic fluid is comparatively a recent one.[13–21] From ESR data one can extract spin-glass correlation, the distribution of internal fields, etc.[22] It is shown that like spin glass, in nanomagnetic systems also the linewidth (ΔH_{pp}) broadens and the resonance field (H_r) shifts towards low field with decreasing temperature.[23–29] Nagata and Ishihara have given a simple relation between the shift of the resonance field and linewidth with temperature.[23] Several other workers have used the concept of spin-glass-like behavior in fine-magnetic-particle systems in order to interpret low-temperature ESR spectra.[25–29] Recently, Koksharov *et al.*[29,30] have studied ESR spectra of iron nitride nanoparticles and shown that below 40 K spectra reveal a sharp line broadening and a shift in the resonance field. The observed linear falloff in the ΔH_{pp} with temperature is explained in terms of the random-field model of exchange anisotropy.

In the present study we analyze results of ESR spectra of fine magnetic particles of $Mn_{0.5}Zn_{0.5}Fe_2O_4$ ferrite dispersed in kerosene. The spectra were recorded at different temperatures. The zero-field-cooled sample exhibits an isotropic shift in resonance field below 50 K while the field-cooled sample exhibits hysteresis in an angular variation of the resonance field. These results are discussed in terms of the thermal dependence of the exchange anisotropy field, which can be described by the random-field exchange anisotropy model. Using this model surface, a spin-glass-like layer of about 3.8 Å thickness is estimated, which agrees with that obtained from small-angle neutron scattering experiments.[31]

II. EXPERIMENTS

The $Mn_{0.5}Zn_{0.5}Fe_2O_4$ nanoparticles were synthesized by a coprecipitation technique followed by digestion. Analytical-grade reagents of ferric, manganese, and zinc chlorides were used to prepare a solution containing Fe^{3+}, Zn^{2+}, and Mn^{2+} ions in the appropriate proportion. This was mixed with an 8 mol NaOH solution, which leads to coprecipitation of ferrite particles. After the precipitation, the suspension was kept for digestion at 90 °C for 30 min. During this time, the particles grew and transformed to the crystalline state. The crystalline nature was confirmed by the x-ray diffraction technique. Oleic acid was then added and the mixture was stirred for an hour. The fluid was heated and peptized by adding a small amount of dilute HCl. By magnetic sedimentation the oleic-acid-coated particles were separated. The suspension containing oleic-acid-coated particles was repeatedly washed with double-distilled water and subsequently washed with acetone to remove the water. Ultimately, this acetone wet

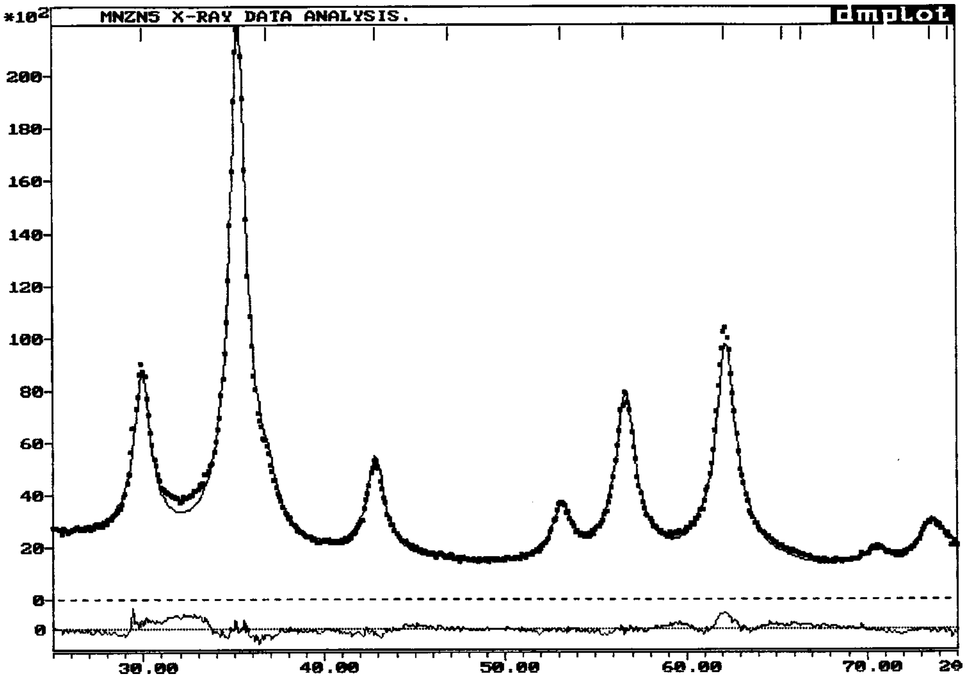

FIG. 1. X-ray diffraction pattern of Mn-Zn ferrite particles. The solid curve is the results of fitting using the Rietveld procedure.

slurry was dispersed in kerosene and acetone was removed by heating. The fluid thus obtained was stable and turns into solid below 200 K.

A. Chemical analysis

The chemical composition of mixed ferrite was confirmed by atomic mass absorption spectra. The resonance wavelengths used were λ (Fe) = 2483 Å, λ (Mn) = 2796 Å, and λ (Zn) = 2193 Å. The ratios of Fe/Mn, Fe/Zn, and Mn/Zn were found to be 3.9 ± 0.1, 3.9 ± 0.1, and 1.0 ± 0.1, respectively, which are very close to the theoretical ratios for respective ions (4, 4, and 1, respectively). In order to prevent agglomeration, particles were coated with oleic acid during synthesis. The presence of coating was confirmed by IR spectroscopy.

B. Structural analysis

The structure of the particle was characterized using an x-ray powder diffractometer. Figure 1 shows the x-ray diffraction pattern of the particle. The pattern was analyzed using Rietveld refinement program.[32] The analysis confirms the formation of single-phase cubic spinel structure. The particle size was determined by Scherrer's formula for (311) reflection. The average crystalline size (diameter) and the lattice parameter thus obtained were 67 Å and 8.450 ± 0.001 Å, respectively.

The morphology of the particle was confirmed by transmission electron microscopy images. The particles were almost spherical with an average diameter of 68 Å.

C. Magnetization measurement

The room-temperature magnetization curve for the magnetic fluid sample is shown in Fig. 2. The system exhibits zero remenance and coercivity. Assuming the log-normal size distribution and using Langevin's theory, the magnetization of such fluid is given by

$$M=\int_0^\infty M(D)P(D)d(D), \tag{1}$$

where

$$M(D)=M_s^1[\coth(\alpha)-1/\alpha], \tag{2}$$

$$\alpha=(M_d HV)/kT.$$

Here V is the volume of the particle, H is the applied magnetic field, k is the Boltzmann constant, and M_d is the domain magnetization of the particle. M_s^1 is the fluid magnetization, which is related to the domain magnetization by

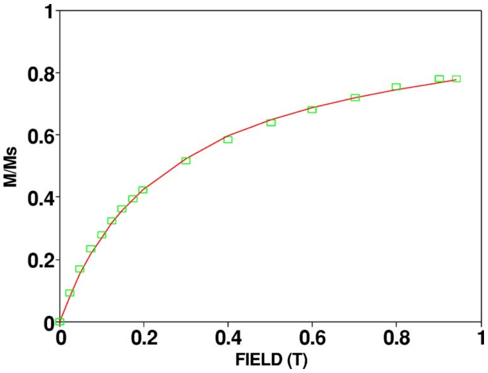

FIG. 2. Room-temperature magnetization curve for the magnetic fluid sample. The solid line is fit to Eq. (1) for $D_0 = 67$ Å, $\sigma = 0.36$, and $M_d = 210$ emu/cc.

$$M_s^1 = M_d \phi, \tag{3}$$

where ϕ = magnetic volume fraction and

$$P(D)d(D) = [D\sigma(2\pi)^{1/2}]^{-1} \times \exp[-\{\ln(D/D_0)\}^2/2\sigma^2]d(D), \tag{4}$$

where σ is the standard deviation of $\ln(D)$, D is the diameter of the particle, and $\ln(D_0)$ is the mean value of $\ln(D)$. The solid line in Fig. 2 is the best fit to Eq. (1) obtain with $D_0 = 67$ Å, $\sigma = 0.36$, and $M_d = 210$ emu/cc. This value of M_d at room temperature is lower than that of the bulk value for the system. The reduced value of magnetization is a characteristic of small particles.[33–40,8] In our earlier publication[31] using small-angle neutron scattering (SANS) experiments it was found that in this system there exists a 3-Å-thick surface layer on the particle, which does not contribute to the magnetic scattering. Taking this thickness into account, the domain magnetization was recalculated and found to be 278 emu/cc, which is close to that of the bulk [283 emu/cc (Ref. 41)].

III. ESR MEASUREMENTS

ESR measurements were carried out using the Bruker-ESR spectrometer operating at an X-band frequency (9–10 GHz). The original fluid has a volume fraction of 6% and was diluted 100 times in order to decrease the interparticle interaction as well as to prevent the radio frequency field line distortion.

ESR measurements were carried out for the ZFC sample and for the FC sample. For the ZFC sample the system was first cooled in zero magnetic field from room temperature to 4.2 K and then ESR patterns were recorded by scanning the field. In order to freeze the position of the particles and to texture the medium for the FC sample, the system was first cooled to 4.2 K in a dc magnetic field of 10 kOe. The spectra were then recorded during th e warming cycle. The angle-dependent ESR spectra were recorded by rotating the Dewar insert containing the FC sample at various angles with respect to the applied magnetic field direction.

At low temperature, the observed linewidth (ΔH_{pp}) is large (>1500 Oe); therefore, one has to take into account the relaxation phenomenon in the analysis. Considering the Landau-Lifshitz formulation and assuming that the observed pattern is due to the superpositions of the microwave absorption of identical ellipsoidal particles (i.e., with the same form factor, magnetization, and relaxation factors), one obtains[42]

$$H_{\mathrm{res}} = H_{\mathrm{res}}^1 + \frac{3}{4}\left(\frac{\Delta H_{\mathrm{pp}}^2}{H_{\mathrm{ref}}}\right). \tag{5}$$

Here H_{res}, H_{res}^1, and H_{ref} are, respectively, the resonance fields without relaxation effect, with relaxation effect, and reference resonance field. In the present case the reference resonance field is 3400 Oe.

Figure 3 show ESR spectra recorded at various temperatures for the zero-field-cooled sample. It is seen that as we warmed the sample the lines sharpen and shift towards the higher field. The variation of resonance field (H_{res}) and ΔH_{pp} with temperature is shown in Fig. 4. The resonance field is corrected using Eq. (5). The behaviors of these parameters are quite distinct in the two regions—viz., (i) $T > 40$ K and (ii) $T \leqslant 40$ K. For $T > 40$ K the resonance field is constant and the linewidth decreases with increasing temperature. For $T \leqslant 40$ K, the resonance field decreases with a decrease in temperature while the linewidth increases rapidly and saturates at about 10 K. A similar behavior was also reported by Koksharov *et al.*[29,30] In the following an attempt is made to explain the above behavior.

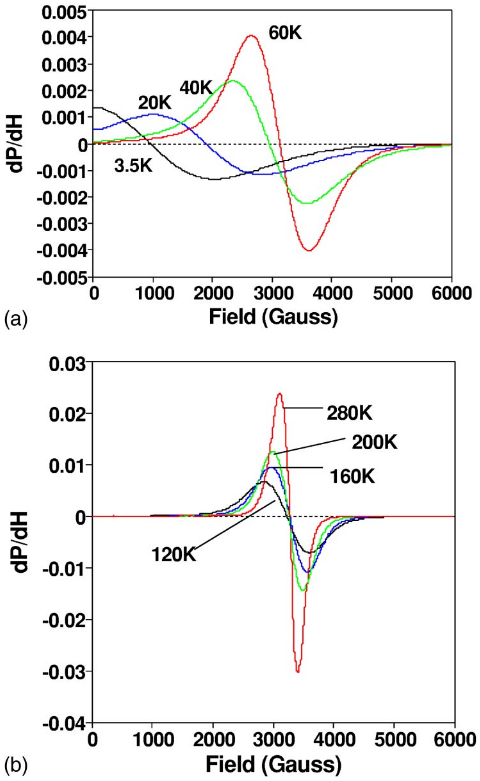

FIG. 3. Typical low-temperature ESR pattern for the dilute magnetic fluid under zero-field-cooled condition—i.e., randomly oriented easy axis.

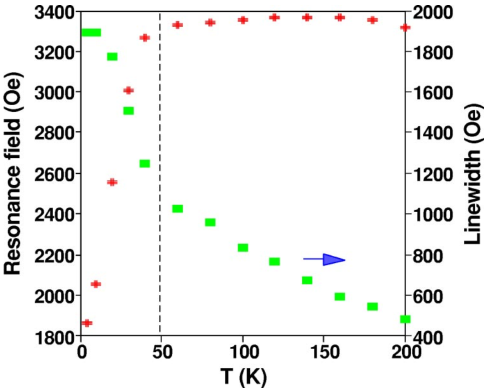

FIG. 4. Variation of resonance field (H_{res}) (+), after correcting for the relaxation effect using Eq. (5), and linewidth (H_{pp}) (□) with temperature for a randomly oriented easy axis sample. The vertical dotted line shows the two different regions (see text).

(i) For $T > 40$ K. According to magnetic resonance theory,[43] the resonance condition for ferrimagnetic and ferromagnetic particles with uniaxial anisotropy is given by

$$H_{\mathrm{res}} = H_{\mathrm{ref}} - H_a P_2(\cos\theta), \tag{6}$$

where $H_a =$ anisotropy field, $P_2(\cos\theta)$ is the second-order Legendre polynomial, and θ is the angle between the axis of the particle and field. According to Eq. (6) for a completely aligned system, H_{res} for parallel (i.e., $\theta = 0°$) and perpendicular (i.e., $\theta = 90°$) configurations is given by

$$H_{\mathrm{res}}(\parallel) = H_{\mathrm{ref}} - H_a \tag{7}$$

and

$$H_{\mathrm{res}}(\perp) = H_{\mathrm{ref}} + \tfrac{1}{2}(H_a). \tag{8}$$

Thus, for the randomly oriented system, the average—i.e., $\langle[\{2H_{\mathrm{res}}(\perp) + H_{\mathrm{res}}(\parallel)\}/3]\rangle$—is equal to H_{ref}, which is constant. This deduction agrees well with the observed constant value of the resonance field for $T > 40$ K.

(ii) For $T \leqslant 40$ K. At temperature below 40 K, $H_{\mathrm{res}}(T)$ decreases while the linewidth increases with decrease in temperature. In a large number of nanomagnetic particle systems such behavior is observed.[25–29] It has been observed from the magnetization study that the strength of the dipole-dipole interaction in certain magnetic fluids is of the order of 40 K.[3,4] Hence dipole-dipole interactions may be responsible for the observed effect in the present study. Considering the value of the magnetic moment of the particle obtained from magnetization data and the volume fraction (i.e., $\mu = 9.1 \times 10^{-20}$ Å m^2), the interparticle distances r for the original and dilute fluids are obtained as 16.3 and 65 nm, respectively. These values give the magnitude of the dipolar interaction between two neighboring particles $[E_d = (\mu_0/4\pi)\mu^2/k_B r^3$, where μ_0 is absolute permeability and k_B is the Boltzmann constant] in terms of temperature as 13 and 0.2 K, respectively for the original and diluted fluids. Therefore, the observed shift in $H_{\mathrm{res}}(T)$ with decreasing T or an abrupt change in linewidth just below 40 K does not seem to be caused by dipole-dipole interactions. Further, in the present system, these observed effects cannot be accounted by Nagate-Ishihara[23] theory. On the other hand, it has been observed that for a system with a spin-glass transition the ESR linewidth usually starts noticeably broadening below the spin-glass temperature. Therefore, in order to interpret the observed behavior for $T \leqslant 40$ K, we assume with Gazeau *et al.*[27,28] and Koksharov *et al.*[29,30] that the isotropic shift $[H_{\mathrm{iso}} = H_{\mathrm{ref}} - H_{\mathrm{res}}(\mathrm{ZFC})]$ and an additional line broadening occur due to the intrinsic spin-glass state which takes place on the surface layer of an individual particle. This state originates due to interactions between spins and results in an internal magnetic structure.

Meiklejohn and Bean[44] have shown that uniaxial exchange anisotropy can exist in a field-cooled nanomagnetic particle with a single-domain ferromagnetic core and antiferromagnetic surface. Martinez *et al.*[5] have shown that the exchange anisotropy can be characterized by the effective field H_E, which depends on the temperature as

$$H_E(T) = H_E(T=0)[1 - T/T_g], \tag{9}$$

where $T_g =$ spin-glass transition temperature.

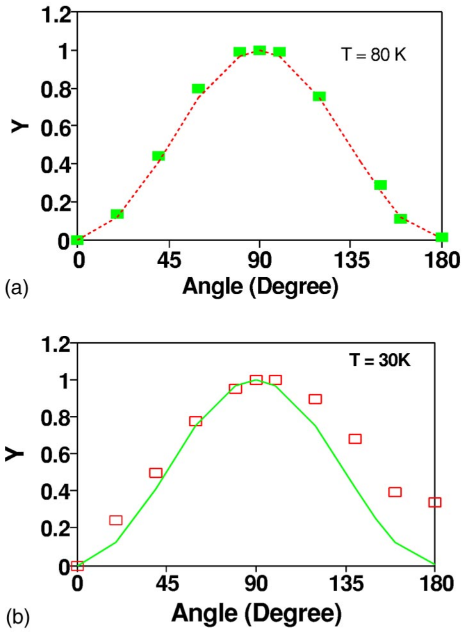

FIG. 5. Angular dependence variation of resonance field at 30 and 80 K for a field-cooled sample. The sample is cooled from 300 K (i.e., in the liquid state) to 4.2 K under a 10-kOe field, where $Y = [H(\theta°) - H(0°)]/[H(90°) - H(0°)]$.

In order to corroborate the above inference ESR patterns were recorded for the field-cooled samples. The fluid was cooled in a 10-kOe field from 300 to 4.2 K. In this situation the anisotropy axis of the majority of the particles will be aligned along the field-cooled direction and the maximum texture will be achieved. At 4.2 K the field was switched off after some time and ESR spectra were recorded for the warming cycle.

Figure 5 shows the angular dependence of the resonance field at 30 and 80 K. The following inferences can be drawn from the figure.

(i) At $T = 30$ K, if we change the angle θ (i.e., the angle between the direction of the field during the cooling and the direction at which sample is positioned while recording the ESR spectrum), the resonance field first increases and reaches a maximum at 90°; thereafter, it decreases and reaches a minimum at 180°. From 180° to 360° (i.e., reversing direction) the H_{res} changes, but does not come back to the original value. Such an irreversible phenomena in ESR may be called an ‘‘angle-dependent hysteresis in resonance field’’ (Fig. 6).

(ii) Further, it was found that at $T = 30$ K, if we again repeat the cycle, the system does not exhibit hysteresis, but follows a $\sin^2\theta$ law given by

$$\frac{H_{\mathrm{res}}(\theta) - H_{\mathrm{res}}(0°)}{H_{\mathrm{res}}(90°) - H_{\mathrm{res}}(0°)} = \sin^2\theta. \tag{10}$$

(iii) For $T = 60$ K the $\sin^2\theta$ law is obeyed in both cases.

Such an angle-dependent hysteresis in the resonance field in a magnetic fluid is not observed earlier.

The following questions may arise from the above observations: (a) At what temperature does this effect disappear? (b) Is this effect a surface effect or a volume effect? (c) What is the origin of this effect?

To answer question (a) ESR spectra for the field-cooled samples were recorded in the following manner. First, the sample was cooled in the field (10 kOe) from 300 to 4.2 K. The field was then switched off. Subsequently, the desired temperature was attained and an angular dependence resonance field was obtained. This procedure was repeated at intervals of 10 K for each temperature from 10 to 80 K. It was found that the hysteresis behavior disappears above 50 K. Thus it may be concluded that some magnetic phase transition is taking place below 50 K.

In order to answer question (b), let us first assume that the hysteresis effect occurs due to spin disorder below 50 K in the whole volume of a particle similar to that in the reentrant spin glass. But it is known that in a reentrant system, the low-temperature spin-glass phase is much sensitive to the externally applied field and the application of a 10-kOe field during cooling makes it disappear. Hence in the present case too similar behavior is expected if the spin disorder is spread throughout the volume of the particle. With this reasoning the following experiments were performed.

(i) First, the fluid was cooled in a 10-kOe field from 300 to 4.2 K.

(ii) The hysteresis behavior was ascertained at 30 K.

(iii) The system temperature was raised to 60 K and the angular dependence resonance field was recorded and found to obey the $\sin^2\theta$ law.

(iv) At this temperature the sample still remains in textured condition and we again cooled the sample from 60 to 4.2 K, but without applying a magnetic field.

(v) But now at 30 K the system followed the $\sin^2\theta$ law rather than the hysteresis behavior observed earlier.

From these observations one may conclude that the hysteresis behavior is not a volume effect. Similarly, it cannot be due to the collective interactions because the hysteresis behavior should reoccur when the FC sample was again cooled from 60 to 4.2 K in zero field, if the dipolar interaction plays a role. Now, if it is assumed that the surface spins freeze below 50 K, then the observed isotropic shift in the ZFC case can be explained as the influence of the exchange anisotropy field on the resonance condition. Following observations further support the above conjunction.

(i) As above, we have cooled the sample from 300 to 4.2 K in a 10-kOe field. The field was switched off ($H = 0$ Oe) and the sample was brought to 60 K.

(ii) At 60 K again we applied the 10-kOe field (it may be noted that the application of 10 kOe at 60 K is not going to affect the core, which is a highly ordered state) and subsequently cooled the sample to 4.2 K.

(iii) At 30 K the system again exhibited hysteresis behavior.

From these experiments it may be concluded that the observed effect is due to surface spin ordering.

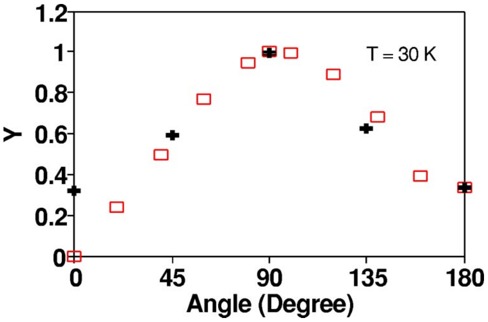

FIG. 6. Angle-dependent hysteresis behavior in magnetic fluid at 30 K for a field-cooled sample. (□, increasing angle: +, decreasing angle), where $Y = [H(\theta°) - H(0°)]/[H(90°) - H(0°)]$.

Recently Chamberlin *et al.*[45] have observed a very unusual feature in their diluted system of monodisperse Fe nanoparticles. They have observed that (i) below the spin freezing temperature relaxation halts abruptly at a nonzero value and (ii) the blocking temperature is inversely proportional to the particle diameter (inconsistent with the Néel-Brown model). This observed behavior is consistent with a type of Casimir-Polder (CP) interaction expected between dilute nanometer-scale particles. In the following we have tried to use a similar concept to explain the observed behavior of the linewidth, resonance field variation in the low-temperature region—i.e., $T \leqslant 50$ K.

ZFC behavior. In the zero-field-cooled condition, surface spin freezes randomly, and if one assumes that a Casimir-Polder interaction (which is attractive) exists between the particle surfaces, then a line broadening (as in the case of the dipolar interaction) and shift in resonance field (i.e., isotropic shift) will be observed. These both effects are observed in the present case (Fig. 4).

FC behavior. In the field-cooled case the surface spins will be frozen, but they will be unidirectional (in the field-cooling direction). Therefore, now the surface-surface interaction is enhanced by directional moment freezing, which will result in line broadening as well as a resonance shift, but now the effects will be enhanced compared to that in the ZFC case. On rotation an additional coupling between particles in the direction of the cooling field may give rise to a

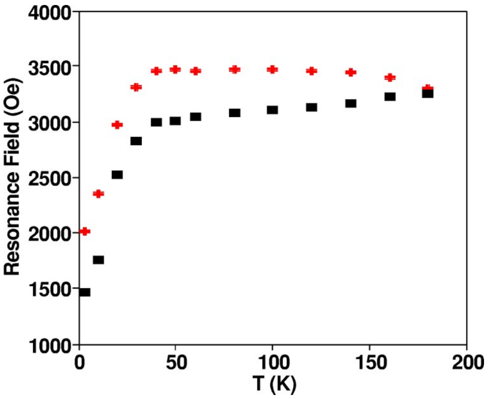

FIG. 7. Resonance field variation with temperature for the textured fluid.

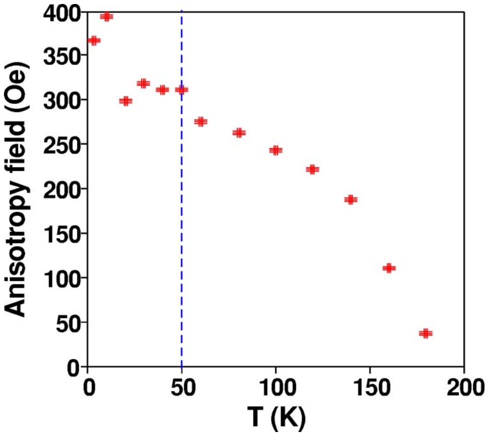

FIG. 8. Thermal variation of anisotropy field (H_a) calculated using Eq. (6), for the textured fluid.

high value of the anisotropy. Accordingly, an abrupt increase in the anisotropy field below 50 K will be observed (Fig. 8 below).

Thus it seems that CP interaction may explain isotropic shift in resonance field, increase in linewidth and increase in anisotropy field below 50 K.

Hysteresis behavior. It is not possible for us to explain the hysteresis effect on the basis of CP interactions. The hysteresis effect was not observed for the FC sample during the experiments (iv) and (v) referred to above. This indicates that the hysteresis is possible only if it is assumed that the surface spins are frozen in a unidirection: i.e., the surface spins are ordered.

Figure 7 shows the variation of resonance field for parallel and perpendicular configurations in the case of the textured fluid. Again two distinct regions are observed. The anisotropy (H_a) was calculated using Eqs. (7) and (8). The H_a monotonically decreases with temperature for $T>50$ K (Fig. 8) while the variation is not smooth below 50 K, indicating that below 50 K a surface spin-glass layer also contributes to the anisotropy. From anisotropy value, H_{ref} values were calculated for the textured magnetic fluid (Fig. 9). The linear fall of $(1-T/T_{\mathrm{crit}})$ is observed for $T_{\mathrm{crit}}=32$ K. If we correlate this shift with the exchange anisotropy field (H_E), then the observed hysteresis behavior on the basis of the model proposed by Martinez *et al.*[5] can be explained.

According to this model, the particle consists of a ferrimagnetic core that changes its orientation by coherent rotation plus a surface spin-glass-like layer that slowly relaxes in the direction of the field. When sample is the ZFC state below 50 K the surface spin-glass layer freezes. This causes a temperature-dependent isotropic shift in the resonance field as well as a temperature-dependent linewidth. In the FC state, a preferred orientation is imposed upon the spin-glass-like surface spins while the FM core remains in a highly ordered state. As soon as the field is removed, the core experiences the field generated by the frozen surface layer in the same direction as that of the previously applied field. This results in hysteresis in $H_r(\theta)$ as well as a shift in H_r. The H_{ref} for the FC state vanishes above 32 K while the freezing temperature of the spin is 50 K. This supports the conjectures that the exchange anisotropy field only exists when the surface spin-glass layer is frozen with a preferred orientation generated with the FC process. This may be the reason why we did not observed hysteresis when we cooled the previously FC sample from 60 to 3.5 K.

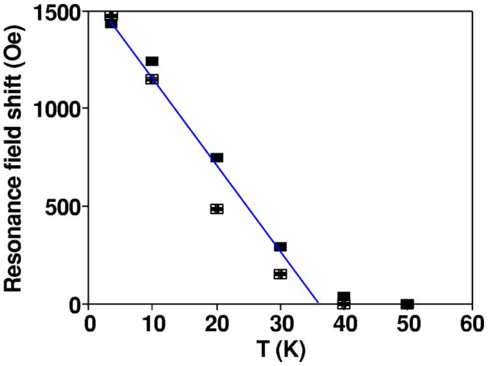

FIG. 9. Resonance field shift (ΔH_{ref}) with temperature for the textured fluid.

Martinez *et al.*[5] have used the theory of an AF/F (antiferromagnetic/ferromagnetic) sandwich[46] to calculate the surface spin-glass layer thickness using exchange anisotropy field values obtained from the experiment and the relation

$$H_E=\frac{2(A_A K_A)^{1/2}}{M_F t_F}, \tag{11}$$

where K_A and A_A are the uniaxial anisotropy and exchange stiffness of the AF and M_F and t_F are the magnetization and thickness of the ferromagnetic core. Using above theory, we have calculated the surface spin-glass layer thickness. In our calculation we have used the value of A_A as 5×10^{-7} erg/cm (same as used in Ref. 5) and other values are $K_A=8.4\times10^4$ erg/cc, calculated from the experimental value of anisotropic field, M_F (at 4.2 K) $=7285$ G,[31] $H_E=1500$ Oe (Fig. 9), thickness of 3.8 Å is obtained which agrees well with the SANS results (3.0 Å).[31]

IV. SUMMARY

Using the ESR technique, the surface spin-glass-like behavior is demonstrated in a textured ferrimagnetic fluid containing nanomagnetic particles of Mn-Zn ferrite. The ZFC sample exhibits an isotropic shift in the resonance field below 50 K while the FC sample exhibits hysteresis behavior in $H_r(\theta)$. The data were analyzed on the basis of existing models.

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